

Study of Low-energy States of Clusters of Spin-1/2 and Spin-1 Triangles with Kagome-like Geometries

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We study the low-energy properties of Heisenberg antiferromagnetic spin-1/2 and spin-1 systems on various clusters made up of triangles. Some of the clusters have a geometry similar to representative pieces of the Kagome lattice, while others have the geometry of a sawtooth chain. While the ground state always has the lowest possible spin (0 or 1/2), the nature of the low-energy excitations depends on the geometry and the site spin. For the Kagome clusters with spin-1 sites, the lowest excitations are gapped, with singlet and triplet excitations having similar gaps. This is in contrast to Kagome clusters with spin-1/2 sites where there are several low-energy singlet excitations (possibly gapless in the thermodynamic limit), while triplet excitations have a gap. For the sawtooth chain with spin-1 sites, the lowest excitations are triplets with a gap; the gap to singlet excitations is about twice the triplet gap.

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There has been a great deal of interest in recent years in quantum spin systems which are strongly frustrated¹. Examples of such systems include lattices involving triangles of Heisenberg spins as motifs which interact antiferromagnetically with each other. Some of the systems which have been studied experimentally in one and two dimensions are the sawtooth chain² and the Kagome lattice³. A variety of theoretical techniques, both analytical and numerical, have been used to study models of such systems⁴⁻¹¹.

Classically (i.e., in the limit in which the site spin $S \rightarrow \infty$), the sawtooth and Kagome antiferromagnetic systems have an enormous ground state degeneracy arising from local rotational degrees of freedom which cost no energy; this leads to an extensive entropy at zero temperature. Quantum mechanically, this degeneracy is lifted due to nonzero matrix elements between different classical ground states. However, one might still expect a remnant of the classical degeneracy in the form of a large number of low-energy excitations. In the quantum limit, it is well-known that the physics of some spin-1/2 and spin-1 lattices in low dimensions is different in nontrivial ways. It is therefore useful to study the lattices involving triangular motifs for site spin-1/2 and spin-1 to see if there is any systematic difference between the two cases.

The spin-1/2 Kagome system is known to exhibit a variety of unusual low-energy properties^{6,7}. For instance, it is believed that there is a small gap between the singlet ground state and the nonsinglet excited states. Secondly, there is a band of singlet excited states with no band gap to the ground state. Finally, the two-spin correlation function decays rapidly with distance indicating that the magnetic correlation length is comparable to the lattice spacing. All these features can be explained qualitatively by assuming that a set of disjoint triangles are in their ground states with total spin-1/2. Then these spin-1/2 states of nearest neighbor triangles can form dimers, thus producing a singlet state¹⁰. The number of such dimer configurations is about 1.15^N if the number of sites is N ; this number is macroscopically large even though it is much smaller than the number of singlets which one can form out of N spin-1/2 objects which is about 2^N . The gapless band of singlet excitations can be thought of as arising from these dimer states, with a small dispersion being produced by the admixture amongst the different dimer states. In contrast to the spin-1/2 case, the ground state of a single spin-1 triangle is unique with a gap to all its excited states. Following arguments similar to the one given above for the spin-1/2 Kagome lattice, one might then expect the ground state of the spin-1 Kagome lattice to be unique with a gap to *all* excitations.

For the spin-1/2 sawtooth chain, the ground state has a degeneracy proportional to the number of sites. This degeneracy arises from the existence of spin-1/2 kinks (a free spin located at the base of a triangle shown in Fig. 6) which do not cost any energy regardless of their lattice position². There are also spin-1/2 antikinks (a free spin located at the top of a triangle in Fig. 6) which cost a finite energy. The lowest excitations in a chain with periodic boundary conditions are given by a kink-antikink pair which has a gap; the total spin of the pair may be either 0 or 1. The spin-1 sawtooth chain had not been studied earlier. We will present a numerical study of this system and will also construct an approximate analytical ground state using a simple physical picture.

In this paper, we will carry out exact diagonalization studies of the Hamiltonian for both spin-1/2 and spin-1 Kagome clusters as well as the spin-1 sawtooth chain. We will discuss the numerical results for the low-energy spectrum in the different cases, and will use trial wave functions to provide an understanding of the results for the spin-1 sawtooth chain.

We will study the lattices of spins shown in Figs. 1-6. Figs. 1-5 show Kagome-type clusters, with the largest number of sites being 21 for spin-1 and 27 for spin-1/2. Fig. 6 shows a 21-site sawtooth lattice which we have studied for site spin-1. The Hamiltonian used in our calculation is the Heisenberg Hamiltonian with isotropic exchange, written as

$$\hat{H} = J \sum_{\langle ij \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \quad (1)$$

where J is the exchange interaction, and $\langle ij \rangle$ sums over the bonded sites shown in Figs. 1-6. We have imposed periodic boundary conditions at the edges of the various clusters as indicated by the dotted lines in Figs. 1-6. The properties of the system can be computed from the eigenstates of the Hamiltonian which are obtained by setting up the Hamiltonian matrix in a suitable basis and diagonalizing it thereafter.

The dimension of the space spanned by the Hamiltonian rapidly increases with the number of spins in the system. Though the use of a constant M_S (i.e., eigenstates of total S^z) basis is quite straightforward, the dimensions of the space for large clusters which have no spatial symmetry are prohibitively large. Hence we have used the valence bond (VB) technique¹² to construct spin adapted functions (eigenstates of the total \hat{S}^2 operator). The advantage of this method is that the dimensions of the constant total S subspaces are much smaller compared to the constant M_S subspaces. After constructing the VB basis, we obtain the Hamiltonian matrix by operating with Eq. (1) on each of the basis states. The matrix representing the Hamiltonian in the VB basis is in general nonsymmetric since the VB basis is nonorthogonal. We use Rettrup's algorithm¹³ to obtain the lowest few eigenstates and eigenvectors in each of the total S subspaces. We have transformed the nonorthogonal VB basis to the orthogonal constant M_S basis which are also eigenstates of the z component of the site spins. This enables us to calculate the spin-spin correlation functions, $\langle S_i^z S_j^z \rangle$, for the different clusters.

For the spin-1 Kagome clusters, we note that the ground state is always a singlet. The lowest excited states are gapped, with the gap for singlet and triplet excitations being approximately equal as shown in Fig. 7. There is a good trial wave function for the singlet ground state due to Hida⁹. We consider each spin-1 as being made out of two spin-1/2's in a symmetrized combination. For the Kagome clusters that we have considered, we see that each spin-1 site belongs to only two hexagons. We can therefore associate each of the composite spin-1/2's (forming a spin-1) with one hexagon. For each hexagon, we then consider the state ψ_0 which is the singlet ground state that arises when six spin-1/2's interact antiferromagnetically with nearest neighbors. A trial wave function for the ground state of the whole cluster is then given by a product of ψ_0 's over all the hexagons, which is then symmetrized between all the pairs of composite spin-1/2's. This trial wave function gives a good estimate of the ground state energy⁹. However, this approximate picture fails to provide a good understanding of the spectrum of low-energy excited states. For six spin-1/2's which form a hexagon and interact antiferromagnetically, the lowest excited state is a triplet and the first singlet excitation lies above this triplet state by about $1.3 J$. However, for the spin-1 Kagome clusters, we find numerically that the singlet and triplet excitations have comparable gaps to the ground state; excitations with spin-2 have a much higher energy. It would be very useful to understand these features of spin-1 Kagome systems, even qualitatively through variational wave functions.

For the spin-1/2 Kagome clusters, we see from Fig. 8 that there is a large number of low-lying states which have the same spin as the ground state. It is possible that in the thermodynamic limit, these states will form a band with the band bottom touching the ground state. On the other hand, the first excited state with spin one higher than the ground state spin seems to be separated from the ground state by a finite gap. All this is in accordance with the numerical results obtained earlier in Ref. [6], and can be understood from the exponentially large number of dimer configurations as mentioned above¹⁰.

We now turn to the two-spin correlation functions $\langle S_i^z S_j^z \rangle$, where i, j denote two sites of a Kagome cluster. Table 1 shows that the correlations fall off very rapidly beyond the second neighbor for the spin-1 case (with 21 sites as in Fig. 3) and beyond the first neighbor for the spin-1/2 case (with 27 sites as in Fig. 5). Thus the magnetic correlation length is very short, although it appears to be a bit longer for the spin-1 Kagome clusters compared to spin-1/2. However, within the second neighbor sites for spin-1 and within the first neighbor sites for spin-1/2, there is no obvious pattern in the correlation functions. For instance, within the triangle 1-2-3 in the spin-1 case (Fig. 3) and the triangle 13-14-15 in the spin-1/2 case (Fig. 5), the three nearest neighbor correlations are not equal. Similarly, in the spin-1 case, the correlation between the second neighbor sites 2-4 is about twice as big as the correlation between the first neighbor sites 3-4. It is possible that these peculiarities are due to finite size effects, which would indicate an unusual sensitivity of the correlation functions to the boundary conditions. It would be very useful to understand these patterns of correlation functions even qualitatively.

Finally, we come to the spin-1 sawtooth chain (Fig. 6). The energies of the low-lying are shown in Fig. 9. We see that there is a gap to the triplet excitation; the singlet excitations have a gap which is about twice as large as the triplet excitation gap. This clearly suggests that the elementary excitations of the spin-1 sawtooth chain are triplets, and the lowest singlet excitations are probably made up of two

well-separated excited triplets.

There is an approximate way of visualizing the ground state and triplet excitations of the spin-1 sawtooth chain. Let us first introduce an algebraic parametrization for spins¹⁴. Let $u = \cos \theta e^{i\alpha}$ and $v = \sin \theta e^{i\beta}$ denote the spin- \uparrow and spin- \downarrow states respectively of a spin-1/2 object. The angles θ, α and β parametrize the surface of the sphere S^3 , lying in the ranges $0 \leq \theta \leq \pi/2$, $0 \leq \alpha, \beta \leq 2\pi$. Then the three states of a spin-1 object are given by

$$\begin{aligned} |S^z = 1\rangle &= \sqrt{3} u^2, \\ |S^z = 0\rangle &= \sqrt{6} uv, \\ |S^z = -1\rangle &= \sqrt{3} v^2, \end{aligned} \quad (2)$$

where the normalization factors on the right hand sides are found by integrating with the measure $d\Omega = d\theta d\alpha d\beta \sin \theta \cos \theta / (2\pi^2)$. (For instance, $\int d\Omega u^{*2} u^2 = 1/3$). Given two spin-1/2 objects at sites i and j , the singlet state $\psi_{i,j}$ is given by $(u_i v_j - u_j v_i)$. Then a trial ground state for the (infinitely long) spin-1 sawtooth lattice is given by

$$\Psi_0 = \prod_{i=-\infty}^{\infty} \psi_{i,i+1}. \quad (3)$$

This is the AKLT state involving all the sites of the chain¹⁵. In this state, the expectation values of $\langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+1} \rangle$ and $\langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+2} \rangle$ are equal to $-4/3$ and $4/9$ respectively¹⁴. For the sawtooth chain, this trial state gives an energy per site, e_0 , equal to $-10/9J \simeq -1.111J$, compared to the numerically obtained value of $-1.117J$. The trial state in Eq. (3) is the unique state which has the maximum number of singlets between the composite spin-1/2 objects (of the site spin-1's) on nearest neighbor sites. It is therefore not surprising that it gives such a good estimate of the ground state energy.

A trial state for the lowest triplet state is given by a superposition of the form

$$\Psi(k) = \sum_n e^{ikn} \left(\prod_{i=-\infty}^{2n-2} \psi_{i,i+1} \right) \psi_{2n-1,2n+1} \left(\prod_{i=2n+1}^{\infty} \psi_{i,i+1} \right), \quad (4)$$

where k denotes the momentum of the state. Each of the wave functions on the right hand side (RHS) of Eq. (4) have the form of a AKLT state which omits the site labeled $2n$ (which lies at the top of a triangle in Fig. 6); it therefore describes a state with total spin-1. It is difficult to compute the expectation value of the Hamiltonian in the state in Eq. (4), particularly because the different states on the RHS are not orthogonal. However, we expect this state to have the lowest energy amongst the triplet trial states because each of the states on the RHS of Eq. (4) is the unique (up to translations) total spin-1 state having the maximum number of singlets between the composite spin-1/2's on nearest neighbor sites. Similarly, a trial state for the next excited state can be constructed as a superposition of AKLT states which omit two spin-1's, say, at sites $2m$ and $2n$ and with wave numbers k_1 and k_2 . The total spin of such states can be either 0, 1 or 2. The bottom of this band may be expected to correspond to a gap which is about twice as large as the gap to the bottom of the band with a single "free" spin-1 excitation. This appears to be the case for the low-energy spectrum of the 19-site spin-1 sawtooth chain shown in Fig. 9; we see that the lowest spin-0 and spin-2 excitations have a gap which is about twice as large as the gap to triplet excitations. For the 21-site chain, we could only get three singlets and two triplets since the Hilbert space of the system is much larger than for the 19-site system. The low-energy spectrum for the states common to the 19-site and 21-site systems are very similar.

To summarize, we have numerically studied a variety of spin-1 and spin-1/2 clusters with triangular motifs. We find significant differences between the low-energy spectra of the spin-1/2 and spin-1 systems. For the Kagome clusters, the spin-1/2 model has a large number of possibly gapless singlet excitations and a finite gap to the lowest triplet excitation. Hence the specific heat goes to zero as a power law

(T^2), while the magnetic susceptibility falls off exponentially as $\exp(-\Delta E_T/k_B T)$ on approaching 0 K⁷. On the other hand, the spin-1 Kagome clusters have a gap to all excitations; hence both the specific heat and the magnetic susceptibility are expected to fall off exponentially as one approaches 0 K. It is difficult to conclude from our studies if these differences are generic to integer spin versus half-odd-integer spin Kagome lattices. For the sawtooth chain, the spin-1/2 model is known to have a number of degenerate ground states; the lowest excitations are singlets and triplets with the same gap². The spin-1 sawtooth chain has a nondegenerate ground state; the lowest excited states are triplets which have a nonzero gap to the ground state. We have presented a simple physical picture of the low-energy states of the spin-1 sawtooth chain. Our numerical results will be useful for developing a quantitative analytical understanding of the low-energy states of the spin-1 Kagome clusters.

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Spin-spin correlation functions for a 21 site spin-1 cluster

Site i	Site j	S=0 Ground State	S=1 Excited State
1	1	0.667	0.656
2	2	0.667	0.793
3	3	0.667	0.656
1	2	-0.171	-0.222
1	3	-0.296	-0.247
2	3	-0.171	-0.222
1	4	-0.021	0.006
1	5	0.076	-0.007
1	6	-0.020	-0.014
2	4	-0.273	-0.180
2	5	0.055	0.150
2	6	0.000	0.030
3	4	0.138	0.078
3	5	-0.136	-0.099
3	6	0.018	-0.005

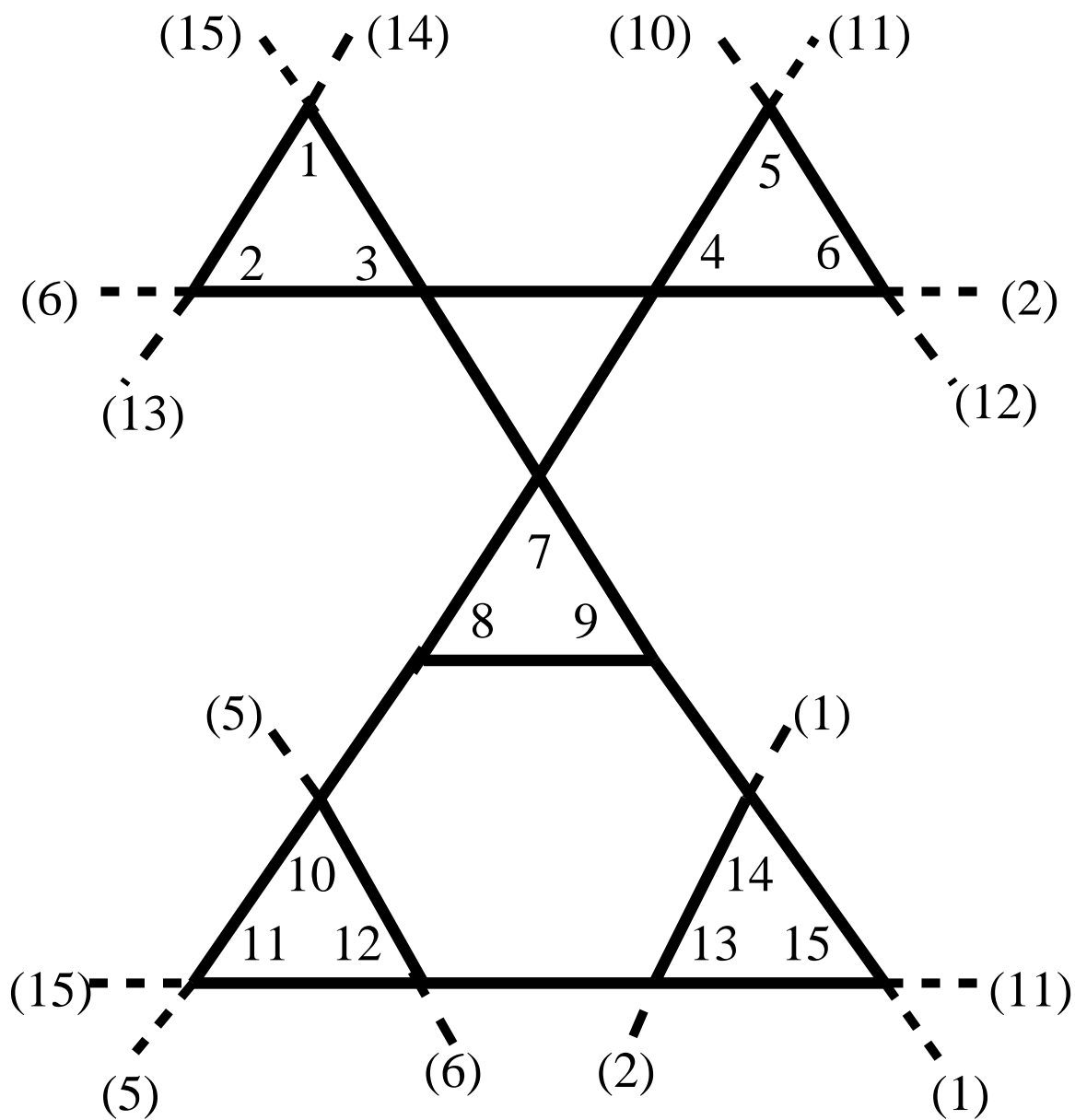
Spin-spin correlation functions for a 27 site spin-1/2 cluster

Site i	Site j	S=0.5 Ground State	S=1.5 Excited State
13	13	0.250	0.250
14	14	0.250	0.250
15	15	0.250	0.250
13	14	-0.128	-0.050
14	15	0.025	-0.107
13	15	-0.128	-0.050
14	10	-0.010	-0.009
14	11	0.003	0.003
14	12	0.002	0.001
13	10	0.002	0.008
13	11	0.004	0.020
13	12	-0.009	-0.009
15	10	0.004	0.004
15	11	-0.012	-0.013
15	12	0.005	0.001

Table 1. Spin-spin correlation functions $\langle S_i^z S_j^z \rangle$ for the 21 site spin-1 Kagome cluster and the 27 site spin-1/2 Kagome cluster.

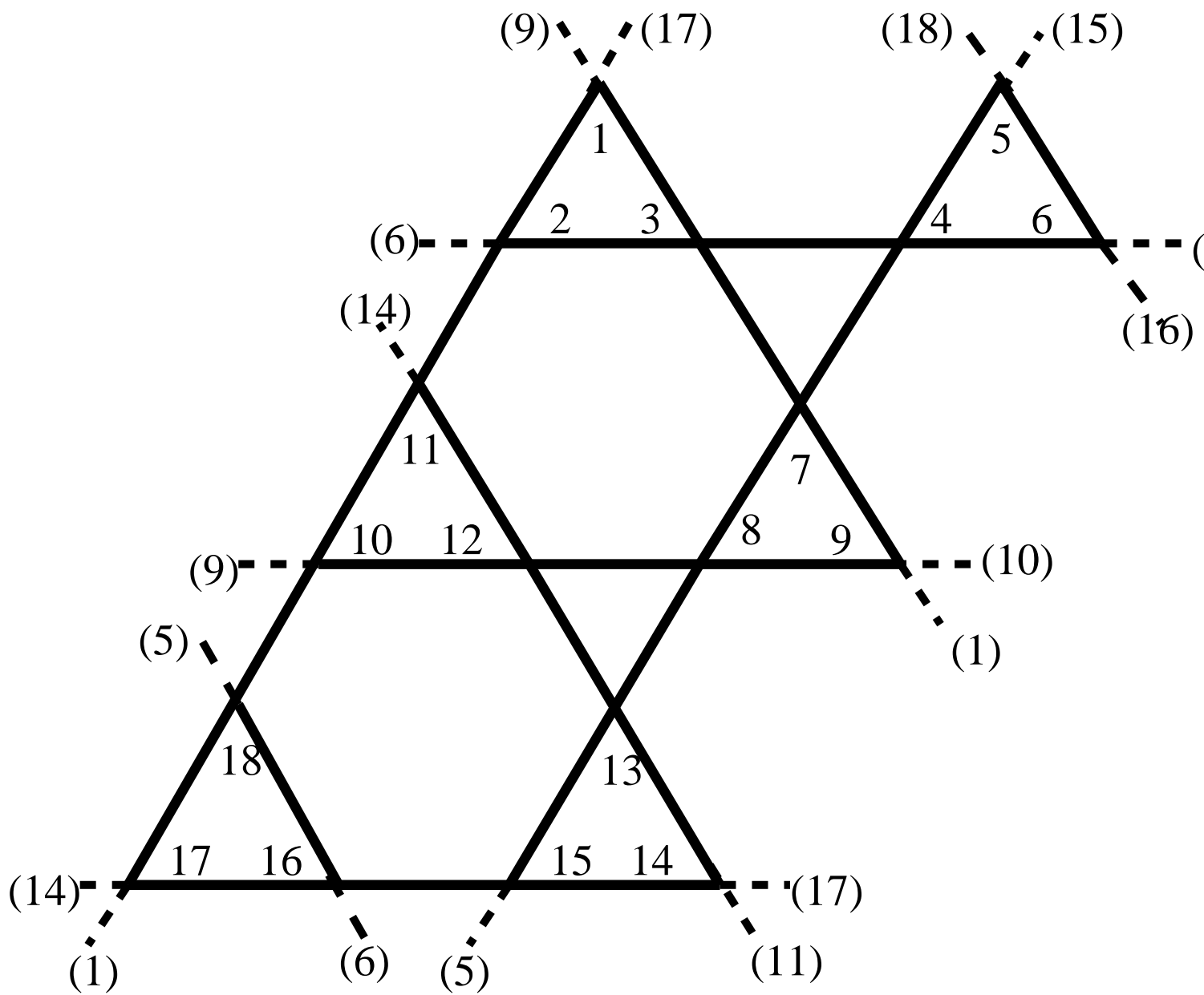
Figure Captions

1. A Kagome cluster of 15 sites.
2. A Kagome cluster of 18 sites.
3. A Kagome cluster of 21 sites.
4. A Kagome cluster of 24 sites.
5. A Kagome cluster of 27 sites.
6. A sawtooth chain of 21 sites.
7. Low-lying energy levels of three spin-1 Kagome clusters (in units of J). The singlet gaps are equal to $0.28 J$, $0.27 J$ and $0.25 J$. The triplet gaps are equal to $0.35 J$, $0.31 J$ and $0.28 J$.
8. Low-lying energy levels of three spin-1/2 Kagome clusters (in units of J). The lowest doublet/singlet gaps are equal to $0.04 J$, $0.04 J$ and $0.04 J$.
9. Low-lying energy levels of two spin-1 sawtooth chains (in units of J). The triplet gaps are equal to $0.25 J$ and $0.24 J$. The singlet gaps are equal to $0.5 J$ and $0.5 J$.



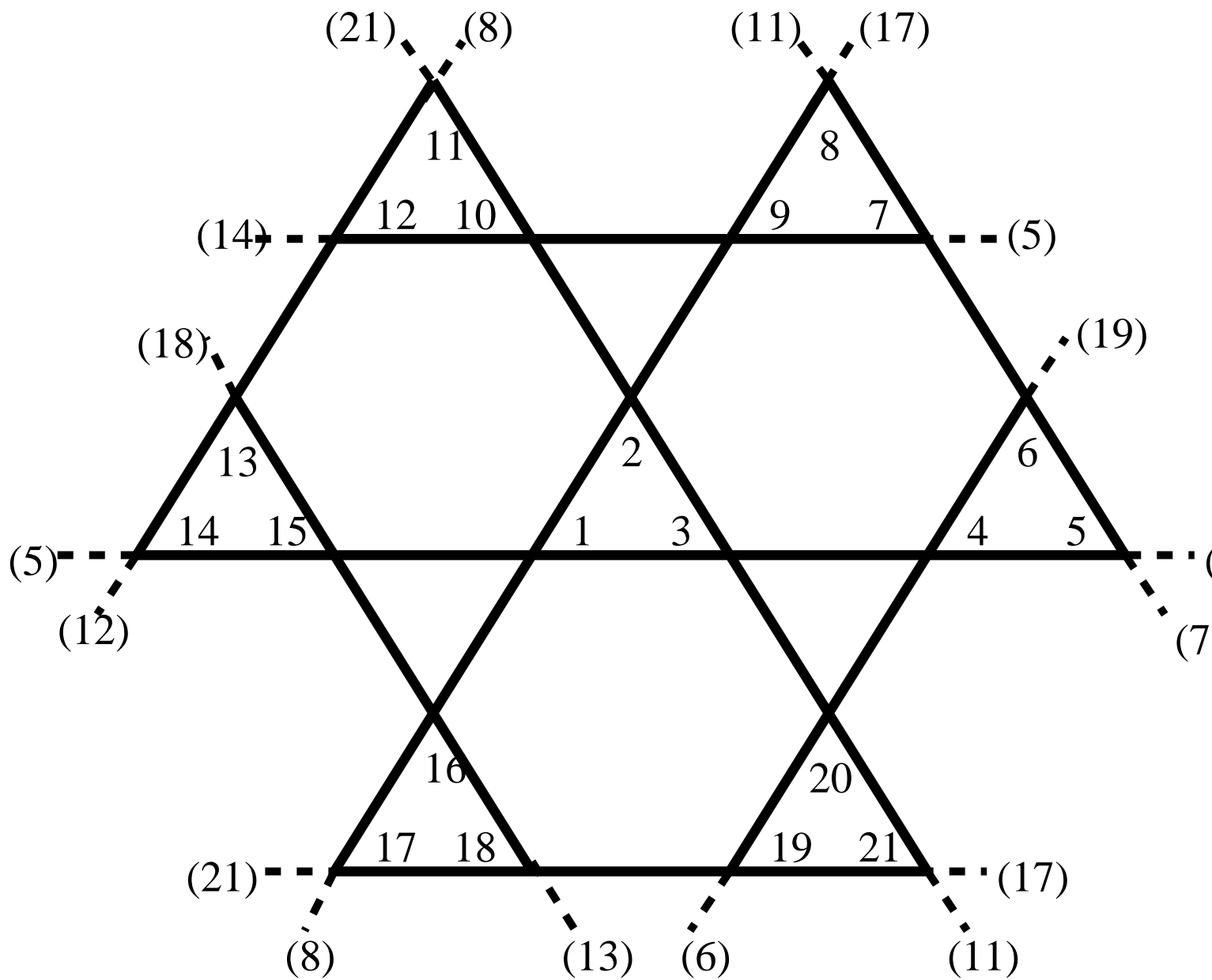
15–Site Strip

Fig. 1



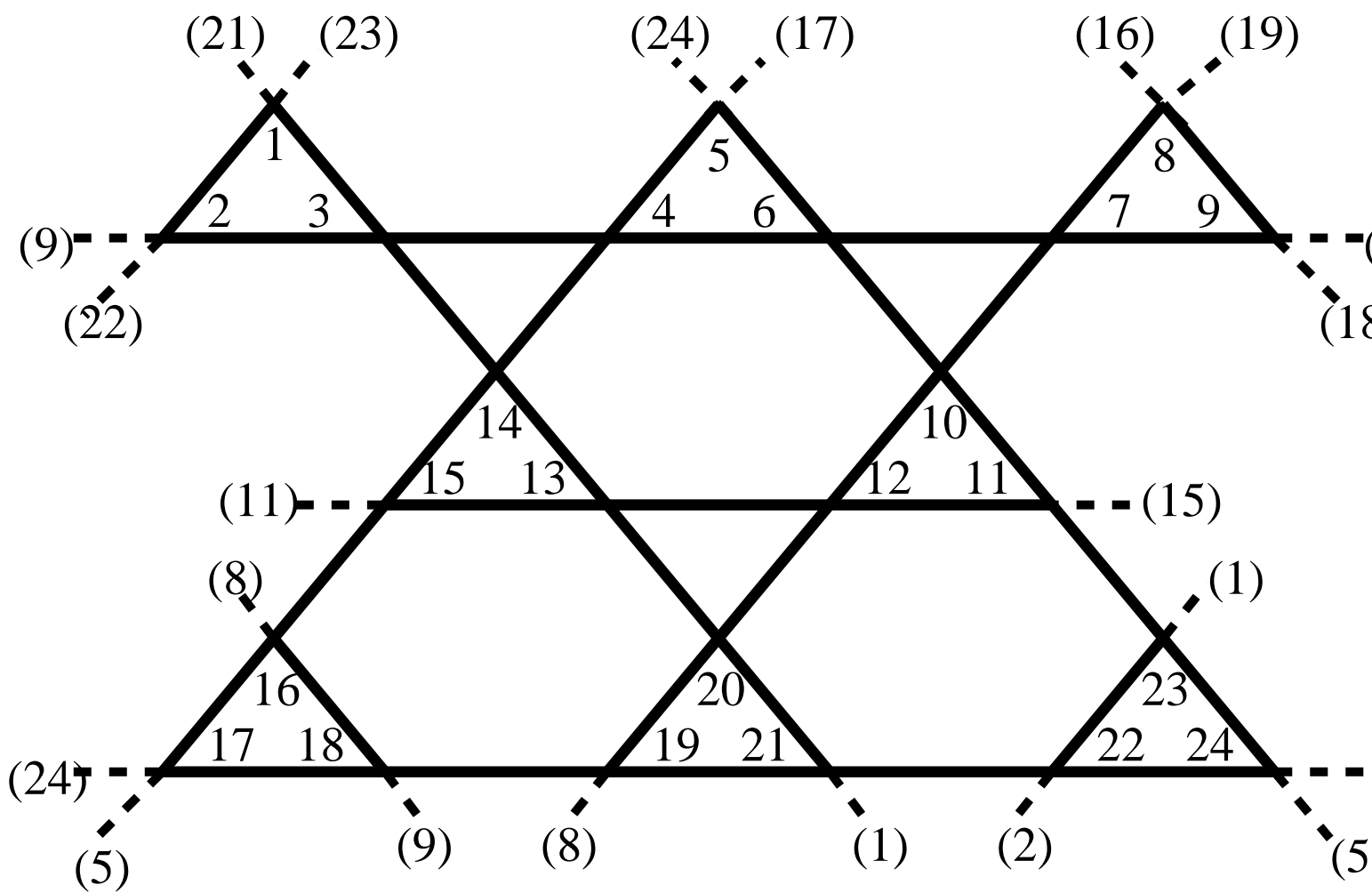
18-Site Strip

Fig. 2



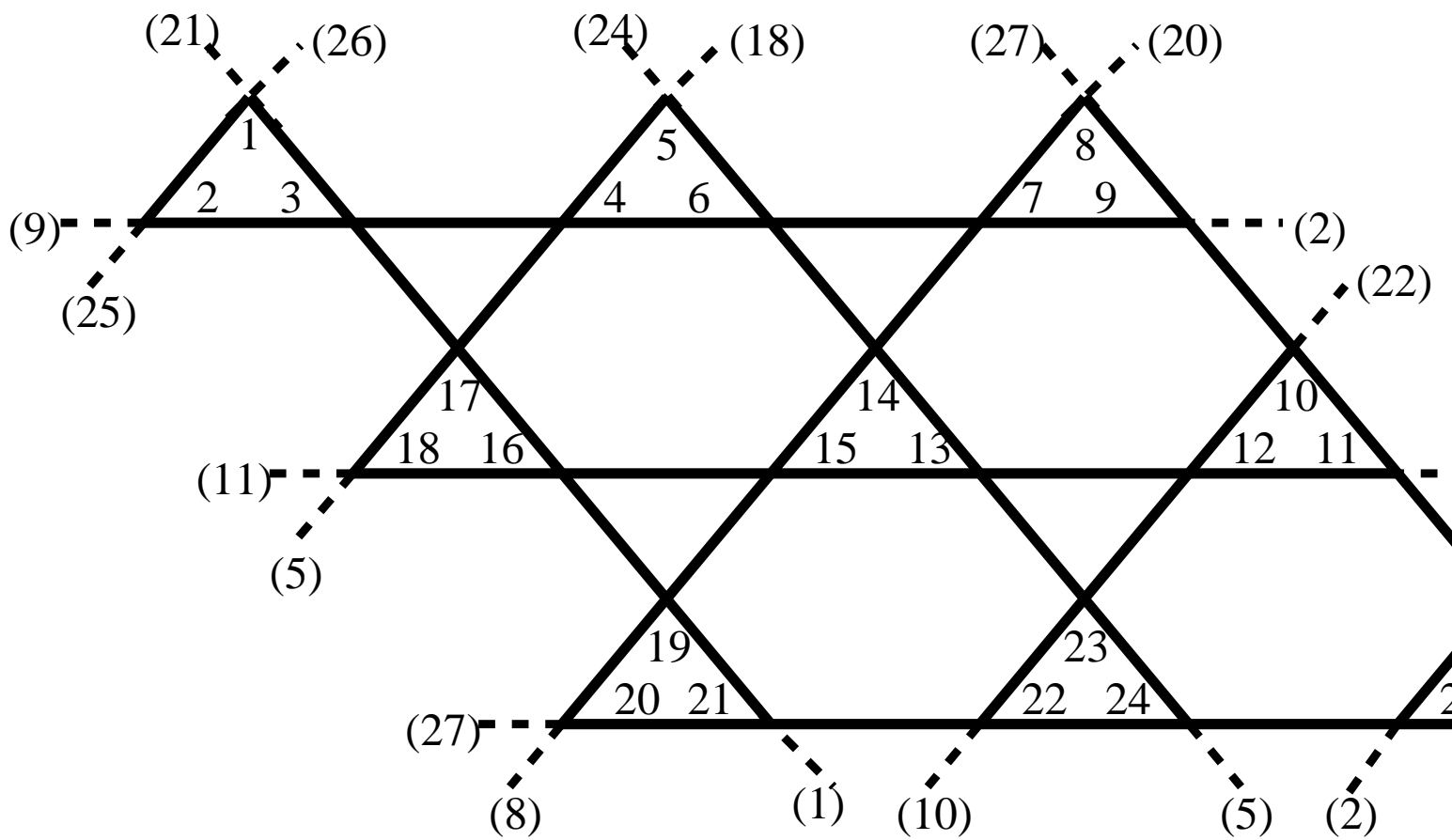
21–Site Strip

Fig. 3



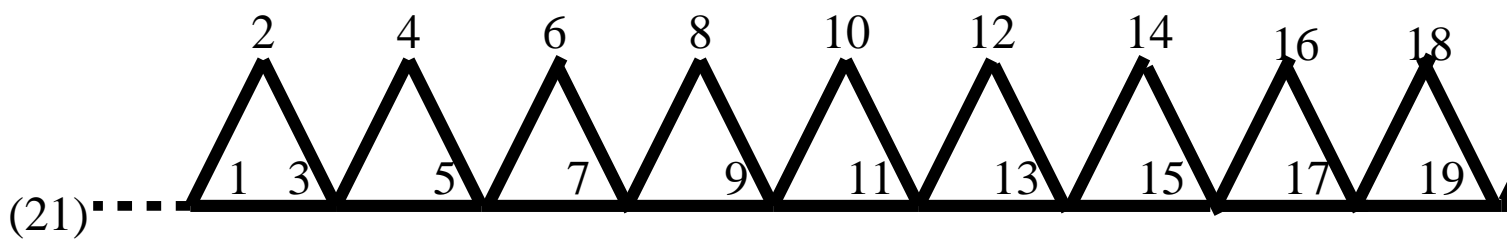
24–Site Strip

Fig. 4



27–Site Strip

Fig. 5



Saw-Tooth Lattice

Fig. 6

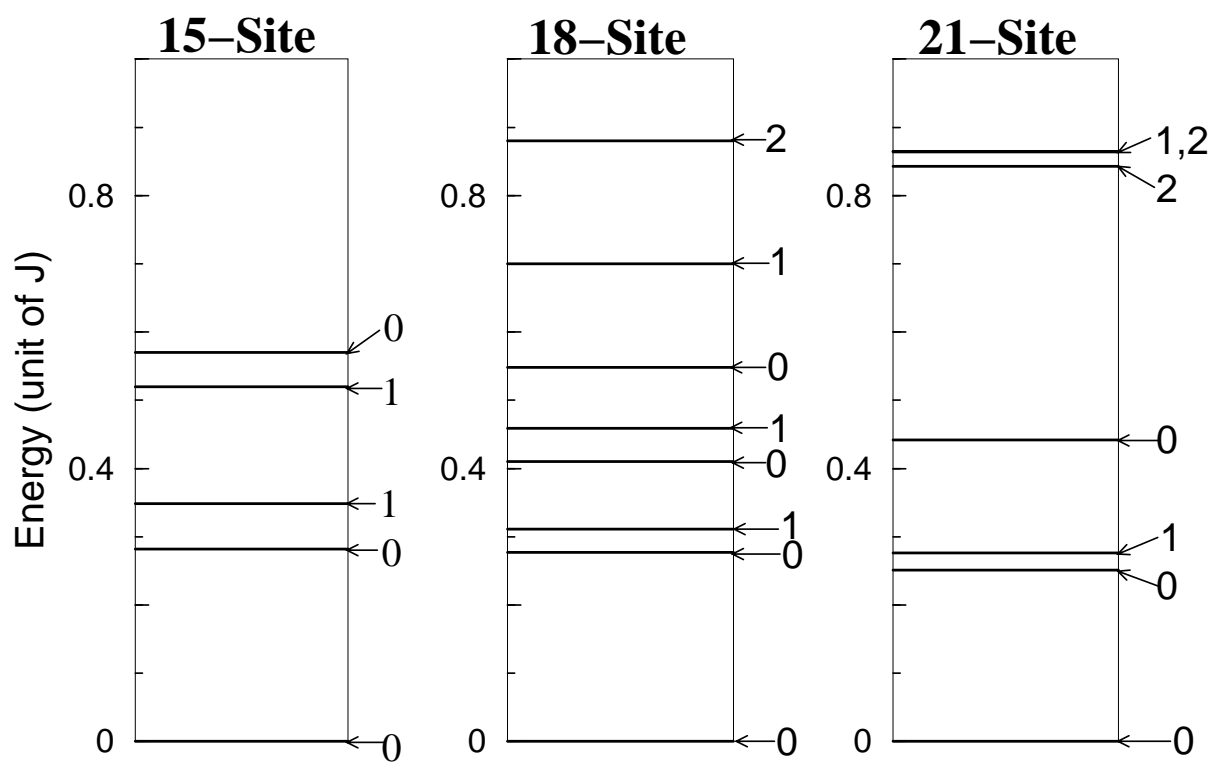


Fig. 7

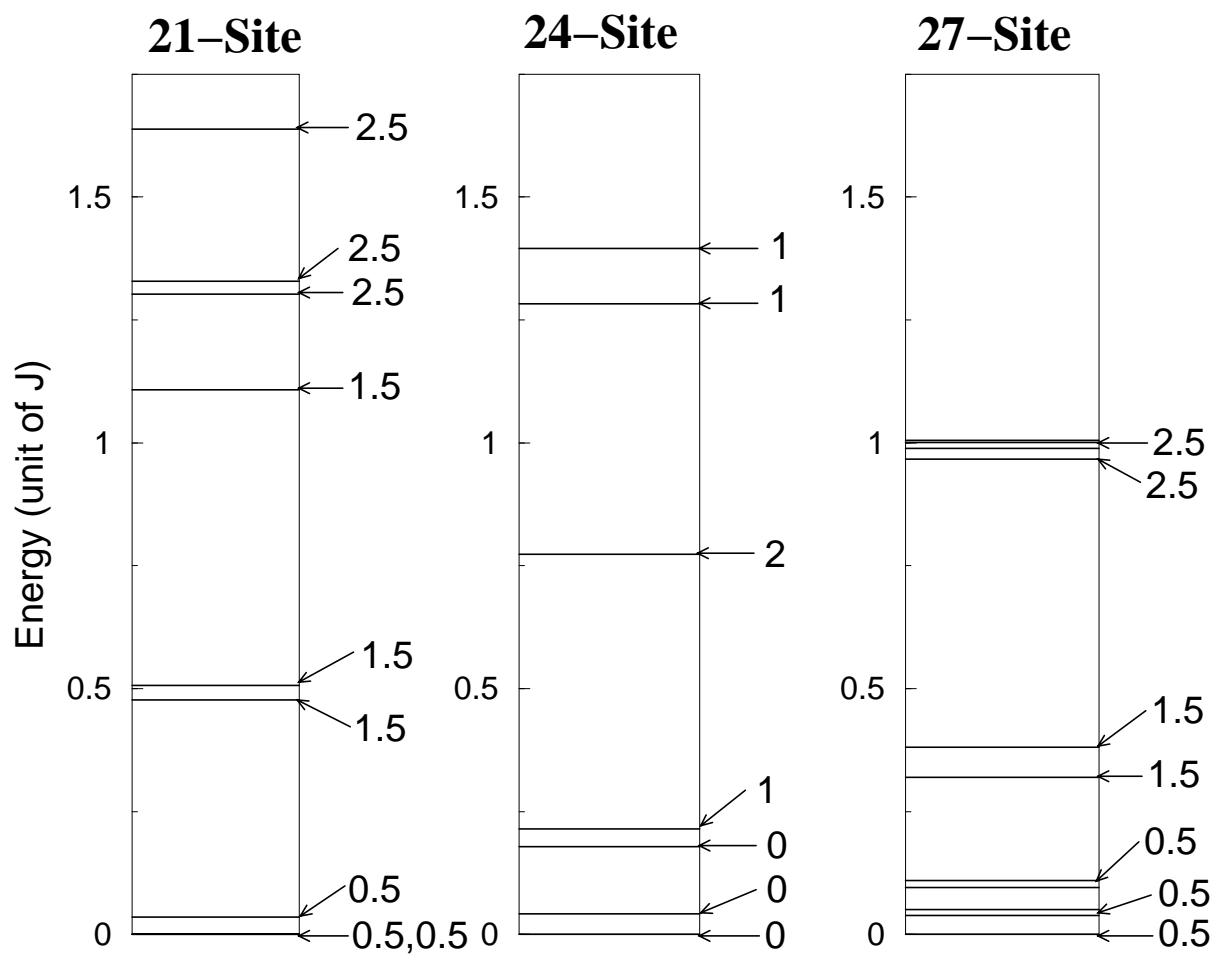


Fig. 8

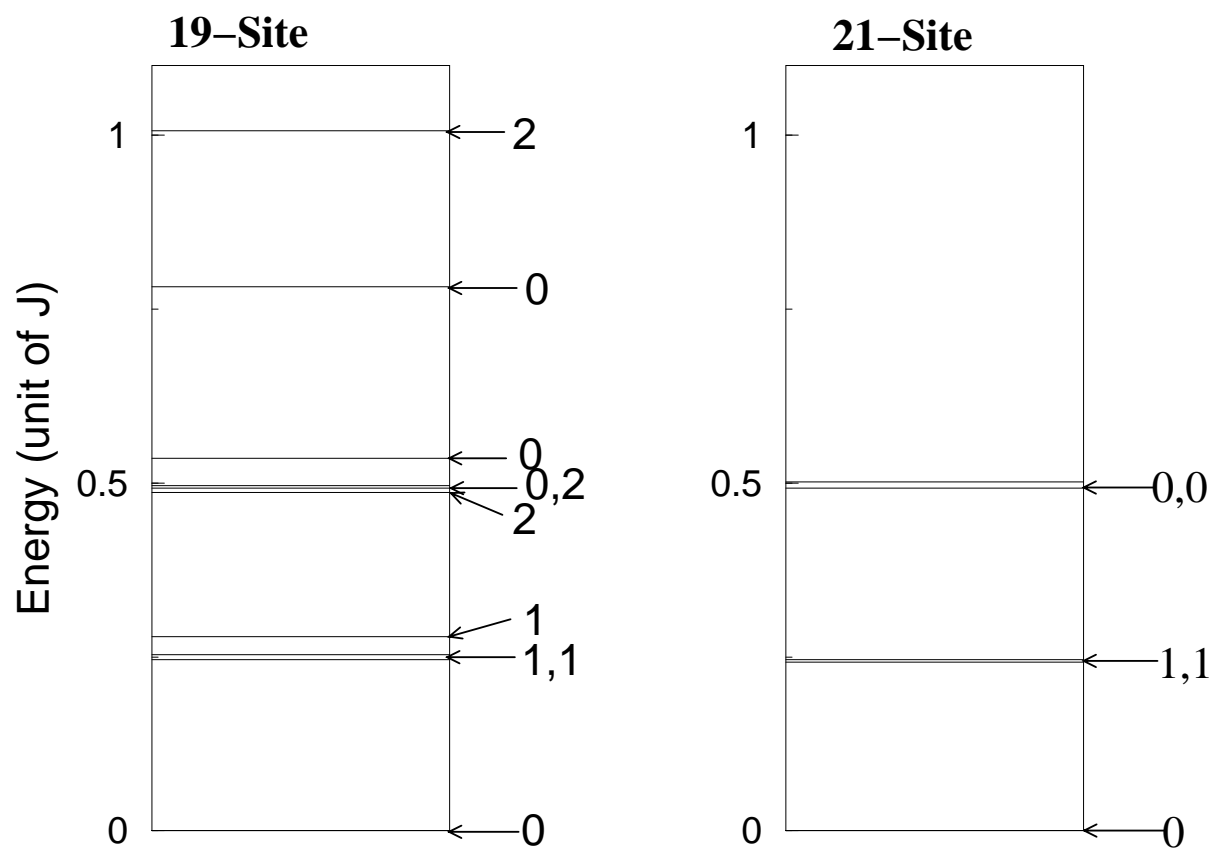


Fig. 9